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Asst. Commissioner for Patents

Washington, DC 20231

OFGS File No. : P/772-274
Inventor : Søren Gyde Thomsen
Title : PROCESS AND REACTOR FOR CARRYING OUT NON-
ADIABATIC CATALYTIC REACTIONS
Assignee : Haldor Topsøe A/S

Enclosed herewith please find the following documents in the above-identified application for United States Letters Patent:

8 Pages of Specification including Abstract and Claims
6 Numbered Claims Calculated as 6 Claims for Fee Purposes
1 Sheet of Drawing Containing Figures 1 to 2. (Formal/Informal)
X Declaration and Power of Attorney
X Priority is Claimed under 35 U.S.C. §119:
Convention Date December 2, 1999 for U.S. Provisional Appln. S.N. 60/168,390

X Certified Priority Application

Small Entity Status is claimed.
X Assignment
X Return-Addressed Post Card

OFGS Check No. 7449, which includes the fee of \$750.00, calculated as follows:

Basic Filing Fee: \$ 710.00
Additional Filing Fees:
Total Number of Claims in Excess of 20, times \$18:
Number of Independent Claims in Excess of 3, times \$80: . .
One or More Multiple Dependent Claims: Total \$270.
Total Filing Fees or
Total Filing Fee Reduced 50% for Small Entity:
Assignment Recording Fee: \$40 40.00
TOTAL Filing Fee and Assignment Recording Fee: \$ 750.00

In the event the actual fee is greater than the payment submitted or is inadvertently not enclosed, or if any additional fee during the prosecution of this case is not paid, the Patent and Trademark Office is authorized to charge the underpayment to Deposit Account No. 15-0700.

Respectfully submitted,



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APPLICATION INFORMATION

Title Line One:: PROCESS AND REACTOR FOR CARRYING OUT

Title Line Two:: NON-ADIABATIC CATALYTIC REACTIONS
Total Drawing Sheets:: 1
Formal Drawings?:: No
Application Type:: Utility
Docket Number:: P/772-274
Secrecy Order in Parent Appl.?:: No

CONTINUITY INFORMATION

This application is a:: NON PROV. OF PROVISIONAL
> Application One:: 60/168,390
Filing Date:: 12-02-1999

Source:: PrintEFS Version 1.0.1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

Søren Gyde THOMSEN

Date: November 28, 2000

Serial No.:

Group Art Unit: Unassigned

Filed: November 22, 2000

Examiner: Unassigned

For: PROCESS AND REACTOR FOR CARRYING OUT NON-ADIABATIC CATALYTIC
REACTIONS

Assistant Commissioner for Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to examination on the merits, please amend the above-identified patent application
as follows:

IN THE SPECIFICATION:

Page 1, before line 1, insert: --This application claims the benefit of U.S. Provisional
Application No. 60/168,390, filed December 2, 1999.

FIELD OF THE INVENTION--;

line 6, insert: -- 2. SUMMARY OF THE INVENTION--.

Page 2, line 3, insert --BRIEF DESCRIPTION OF THE DRAWINGS--;

line 11, insert --DETAILED DESCRIPTION OF THE PREFERRED
EMBODIMENT--.

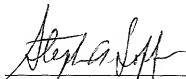
Page 6, line 1, change "CLAIMS" to --WHAT IS CLAIMED IS--.

REMARKS

By this Preliminary Amendment, the specification has been amended to include a reference to the priority provisional application and to add proper headings.

Prompt examination is respectfully requested.

Respectfully submitted,



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16 November 2000

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HABS/AKN

**Process and Reactor for carrying out
Non-Adiabatic Catalytic Reactions**

The present invention relates to a process and reactor system for carrying out non-adiabatic reactions proceeding in a process gas in presence of a catalyst exothermically or endothermically in indirect heat exchange with an appropriate heat exchange medium.

A general object of this invention is thus to provide a process for carrying out non-adiabatic reactions comprising the steps of:

introducing in parallel a first stream of reactants into a first reaction space and a second stream of reactants into a second reaction space,

at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect heat exchange with a heat exchanging medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with the heat exchanging medium, the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space, the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging relationship with the heat exchanging medium.

The invention is in particular useful in carrying out steam reforming reactions in a hydrocarbon feed stock by heat supplied from hot effluent gas from an autothermal steam

reforming reactor and steam reformed product gas from the process.

A specific embodiment of the reaction system according to the invention is described more detailed in the following description by reference to the drawings in which Fig. 1 shows schematically a reaction system being used in the production of a gas with a high content of hydrogen and/or carbon monoxide from steam-reforming of a hydrocarbon feed stock.

Steam reforming is an endothermic chemical reaction, where hydrocarbons and steam react on a steam reforming catalyst, and if appropriate heat is supplied to the location of the reaction.

The reactor system being used in this embodiment consists of three reactors, wherein the steam reforming process is carried through. The three reactors R1, R2 and R3 are operated in parallel.

R1 is an adiabatic reactor. The reactants for the process in R1 consist of hydrocarbon, steam and an oxygen rich gas being introduced into the reactor at an appropriate temperature and mixed. The oxygen and the hydrocarbon will react by combustion and result in a hot gas of residual hydrocarbon, steam and resulting in products from the combustion. Subsequently, the hot gas is passed through a bed of reforming catalyst and catalytically converted to a hot mixture of hydrogen, carbon monoxide and carbon dioxide.

R2 and R3 are two plug flow reactors. The reactants for the process in R2 and R3 are a mixture of hydrocarbon and steam, which is heated to an appropriate temperature before flowing through a bed of reforming catalyst. Walls surround and enclose the catalyst beds of R2 and R3. A hot gas is flowing outside these walls countercurrent to the reacting gases in the catalyst beds. Heat is conducted through the walls from the hot gas to the reacting gases, while the gases are converted to a hot mixture of hydrogen, carbon monoxide and carbon dioxide.

The product gases from R1, R2 and R3 are mixed and form the hot gas flowing outside the walls of R2 and R3, where they form the heat source of the reactions in R2 and R3. This gas is called the heating gas.

As a general advantage of the invention, the walls of R2 and R3 can be arranged in a layout so as to form an optimal channel for the heating gas.

The invention provides, furthermore, a reaction system being in particular useful for carrying out the above process. In general, the reaction system of this invention comprises connected in parallel a first and a second reaction compartment being adapted to hold a catalyst and to receive a reactant stream, the first compartment being in form of a reactor tube, wherein

a first heat exchange space concentric and spaced apart surrounds the first reaction compartment, and the second reaction compartment surrounds a second heat exchange space.

Reactor R2 contains the catalyst inside tubes. Reactor R3 holds the catalyst outside the tubes. The combined reactor R2 and R3 comprises a number of double-tubes, where the inner tubes are catalyst filled (R2) and the double-tubes are in addition arranged in a pattern allowing the volume between the double-tubes to be filled with catalyst as well, i.e. reactor R3. The sensible heat from the combined product gas from the reactors R1, R2 and R3 is cycled back to the reactors R2 and R3. The product gas is flowing in annular channels provided by the double-tubes, counter-currently to the flow in the reactors R2 and R3. Heat is supplied to reactor R2 via the inner wall of the double pipes and the reactor R3 is supplied with heat from the outer wall of the double-tubes.

The advantage of the combined reactor as shown in Fig. 2 is that the heat exchange channels are utilised in an optimal manner, i.e. both the inner wall and the outer wall are utilised as exchange heat surfaces thus making optimal use of expensive material. This also leads to a very compact design of equipment compared to other types of heat exchange reformers and at the same time provides low pressure drop.

On cooling the product gas, a certain risk of metal dusting corrosion exists. A further advantage of the combined reactor design is restricted risk of metal dusting to a limited surface.

The double tube dimensions are typically: Inner tube OD 50 to 140 mm and outer tube OD 80 to 170 mm. The layout can be but need not be arranged in such a way that the heat ex-

change/ area/catalyst volume ratio is equal for the inner tubes and the outer tubes.

CLAIMS

1. Process for carrying out non-adiabatic reactions comprising the steps of:

5 introducing in parallel a first stream of reactants into a first reactions space and a second stream of reactants into a second reaction space;

at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect heat exchange with a heat exchanging medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with a heat exchanging medium, and withdrawing a first and second steam reformed product gas; and

15 the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space, the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging relationship with the heat exchanging medium.

25 2. Process of claim 1, wherein the non-adiabatic reaction is endothermic steam reforming of a hydrocarbon feedstock.

3. Process of claim 1, wherein the heat-exchanging medium comprises an effluent stream from autothermal steam reforming of a hydrocarbon feed stock and/or the product gas.

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4. Reaction system for carrying out non-adiabatic catalytic reactions, comprising connected in parallel a first and second reaction compartment being adapted to hold a catalyst and to receive a reactant stream, the first compartment being in form of a reactor tube, wherein

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a first heat exchange space concentric and spaced apart surrounds the first reaction compartment, and the second reaction compartment surrounds a second heat exchange space.

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5. Reaction system of claim 4, wherein the first and second reaction compartment are arranged within a common shell.

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6. Reaction system of claim 4, wherein the first and second heat exchange space are formed by a common passage-way.

ABSTRACT

Process for carrying out non-adiabatic reactions comprising the steps of:

- 5 introducing in parallel a first stream of reactants into a first reaction space and a second stream of reactants into a second reaction space;
at reaction conditions contacting the first reactant stream with a catalyst in the first reaction space in indirect
- 10 heat exchange with a heat exchanging medium and contacting the second reactant stream with a catalyst in the second reaction space in indirect heat exchange with a heat exchanging medium, and withdrawing a first and second steam reformed product gas; and
- 15 the catalyst in the first reaction space being arranged within a tubular reactor in indirect heat exchanging relationship with the heat exchanging medium by introducing the medium into tubular heat exchange space concentrically surrounding the tubular reactor with the first reaction space,
- 20 the catalyst in the second reaction space being arranged on shell side of a heat exchange space in indirect heat exchanging relationship with the heat exchanging medium.

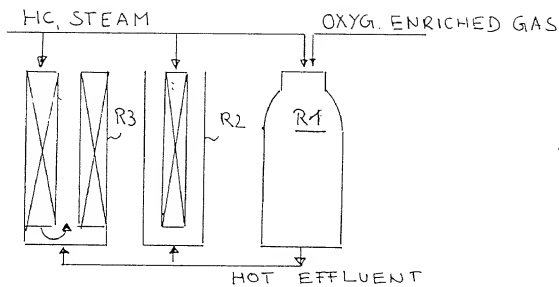


Fig. 1

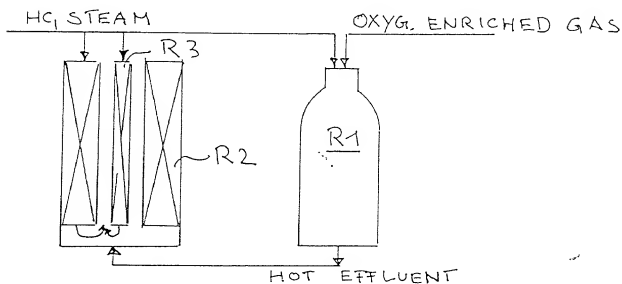
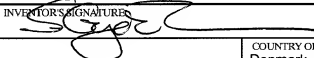


Fig. 2

UNITED STATES OF AMERICA		OFGS FILE NO.	
COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION		P/772-274	
<p>As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I verily believe that I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural inventors are named) of the subject matter which is claimed and for which a patent is sought on the invention entitled:</p> <p>"PROCESS AND REACTOR FOR CARRYING OUT NON-ADIABATIC CATALYTIC REACTIONS"</p>			
<p>the specification of which is attached hereto, unless the following box is checked:</p> <p><input type="checkbox"/> was filed on _____ as United States patent Application Number or PCT International patent application number _____ and was amended on _____ (if any).</p> <p>I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.</p> <p>I acknowledge the duty to disclose all information known to be material to patentability in accordance with Title 37, Code of Federal Regulations, §1.56.</p> <p>I hereby claim priority benefits under Title 35, United States Code §119 of any foreign application(s) for patent or inventor's certificate or United States provisional application(s) listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:</p>			
Prior Foreign or Provisional Application(s)			
COUNTRY	APPLICATION NUMBER	DATE OF FILING <small>(day, month, year)</small>	PRIORITY CLAIMED UNDER 35 U.S.C. 119
U.S.A.	60/168,390	02 December 1999	YES <input checked="" type="checkbox"/> NO <input type="checkbox"/>
			YES <input type="checkbox"/> NO <input type="checkbox"/>
			YES <input type="checkbox"/> NO <input type="checkbox"/>
<p>I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.</p>			
UNITED STATES APPLICATION NUMBER	DATE OF FILING <small>(day, month, year)</small>	STATUS <small>(patented, pending, abandoned)</small>	
<p>I hereby appoint OSTROLENK, FABER, GERB & SOFFEN, and the members of the firm, Marvin C. Soffen - Reg. No. 17,542; Samuel H. Weiner - Reg. No. 18,510; Jerome M. Berliner - Reg. No. 18,653; Robert C. Faber - Reg. No. 24,322; Edward A. Meilmann - Reg. No. 24,735; Stanley H. Lieberstein - Reg. No. 22,400; Steven I. Weisburd - Reg. No. 27,409; Max Moskowitz - Reg. No. 30,576; Stephen A. Soffen - Reg. No. 31,063; James A. FINDER - Reg. No. 30,173; William O. Gray, III - Reg. No. 30,944 and Louis C. Dujmich - Reg. No. 30,625, as attorneys with full power of substitution and revocation to prosecute this application, to transact all business in the Patent & Trademark Office connected therewith and to receive all correspondence.</p>			
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OSTROLENK, FABER, GERB & SOFFEN 1180 AVENUE OF THE AMERICAS NEW YORK, NEW YORK 10036-8403		(212) 382-0700	
<p>I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.</p>			
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RESIDENCE		DATE	
COUNTRY OF CITIZENSHIP			
POST OFFICE ADDRESS			
FULL NAME OF THIRD JOINT INVENTOR (IF ANY)		INVENTOR'S SIGNATURE	
RESIDENCE		DATE	
COUNTRY OF CITIZENSHIP			
POST OFFICE ADDRESS			